MATHEMATICAL MODELING OF THE PROCESS OF FORMATION OF THIN COATINGS BY CENTRIFUGING WITH THE AIM OF DETERMINING AN EFFICIENT REGIME

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UDC 681.3.06

The process of formation of thin coatings by contrifuging is considered; factors that diminish the quality of the produced layers are isolated. A viscous incompressible liquid is analyzed in different regimes of rotation, and the most efficient regime is found.

There are a variety of technological processes in industry that involve applying thin films by the centrifuging method, for example, in electronics production. In investigating these operations one usually considers either just liquid flow with a known law of rotation [1-3] or the resultant film thickness profile [4-6]. However, to produce the most uniform coatings, it is important to reveal the reasons why irregularities form and to determine ways of decreasing them. In this connection the problem of determining an efficient regime of rotation for a plate when it is covered with a viscous incompressible liquid with the aim of ensuring high uniformity of the layer arises.

In constructing a mathematical model we will consider that a continuous plate of radius R rotates around its center with a time-varying angular velocity ω and a viscous incompressible liquid is supplied in the vicinity of the center with the flow rate Q.

In a drop we will isolate the considered volume on the boundary of a resistor dose (Fig. 1), bounded by the angle $d\theta$, of radius r and height h equal to the drop height of the drop. We will consider that this volume moves in the same manner as in a full cylinder of radius r, the entire supplied liquid being uniformly distributed over the surface of the drop. The flow of the volume element of the liquid in this case can be described by a system of Navier-Stokes and continuity equations:

$$\frac{\partial U}{\partial t} + U\frac{\partial U}{\partial r} + W\frac{\partial U}{\partial z} - \frac{V^2}{r} = \nu\left[\frac{\partial^3 U}{\partial r^2} + \frac{1}{r}\frac{\partial U}{\partial r} - \frac{U}{r^2} + \frac{\partial^2 U}{\partial z^2}\right]; \tag{1.1}$$

$$\frac{\partial V}{\partial t} + U\frac{\partial V}{\partial r} + W\frac{\partial V}{\partial z} - \frac{VU}{r} = \nu\left[\frac{\partial^2 V}{\partial r^2} + \frac{1}{r}\frac{\partial V}{\partial r} - \frac{V}{r^2} + \frac{\partial^2 V}{\partial z^2}\right]; \tag{1.2}$$

$$\frac{1}{r}\frac{\partial (rU)}{\partial r} + \frac{\partial W}{\partial z} = 0. \tag{1.3}$$

We should add to them the mass balance equation (the increase in the liquid mass dm on the plate surface in the time dt):

$$dm = Qdt. \tag{1.4}$$

Using the vanishing of the tangential stresses on the liquid surface, the kinematic condition, and the vanishing of the radial and normal velocity components and the equality of the tangential component to ωr on the plate surface, the boundary conditions can be determined as

Voronezh Technological Institute. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 66, No. 5, pp. 561-567, May, 1994. Original article submitted August 10, 1992.

1062-0125/94/6605-0496$12.50

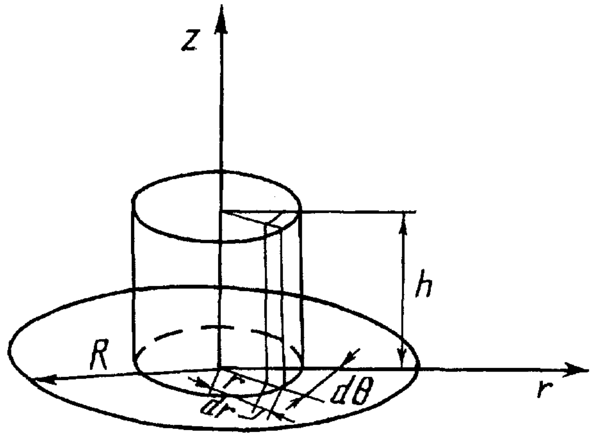

Fig. 1. Calculation scheme.

$$z = 0: \; U = W = 0; \; V = \omega r; \tag{2.1}$$

$$z = h: \; \frac{\partial U}{\partial z} = \frac{\partial V}{\partial z} = 0; \quad W = \frac{\partial h}{\partial t} + U\frac{\partial h}{\partial r}, \tag{2.2}$$

and the initial ones on the upper boundary ($z = h$) as

$$t = 0: \; U = U_0; \; V = 0; \; W = W_0, \tag{3}$$

which are due to the outflow of the liquid from the batcher without rotation about a vertical axis.

Since flow of the liquid occurs over a rotating plate, for analyzing the hydrodynamics of spreading it makes sense to pass to relative motion whose velocity components are rewritten as

$$U = U; \; V = V - \omega r; \; W = W. \tag{4}$$

The system of equations (1) and boundary conditions (2) will be transformed in the following manner:

$$\frac{\partial U}{\partial t} + U\frac{\partial U}{\partial r} + W\frac{\partial U}{\partial z} - \frac{V^2}{r} = \nu\frac{\partial^2 U}{\partial z^2} + 2\omega V + \omega^2 r; \tag{5.1}$$

$$\frac{\partial V}{\partial t} + U\frac{\partial V}{\partial r} + W\frac{\partial V}{\partial z} + \frac{VU}{r} = \nu\frac{\partial^2 V}{\partial z^2} + 2\omega U + r\frac{d\omega}{dt}; \tag{5.2}$$

$$\frac{V}{r} + \frac{\partial U}{\partial r} + \frac{\partial W}{\partial z} = 0; \tag{5.3}$$

$$dm = Qdt; \tag{5.4}$$

$$z = 0: \; U = V = W = 0; \tag{5.5}$$

$$z = h: \; \frac{\partial U}{\partial z} = \frac{\partial V}{\partial z} = 0; \quad W = \frac{\partial h}{\partial t} + U\frac{\partial h}{\partial r}. \tag{5.6}$$

We will seek the solution of the system as

$$U = \sum_{i=1}^{\infty} a_i(t)\, \omega r \delta^i; \quad (6.1)$$

$$V = \sum_{i=1}^{\infty} b_i(t)\, \omega r \delta^i; \quad (6.2)$$

$$W = -\sum_{i=1}^{\infty} 2a_i(t)\, \omega h \frac{\delta^{i+1}}{i+1}. \quad (6.3)$$

Since we consider that the law of velocity distribution over the thickness is parabolic, after substituting (6) into system of equations (5) and integrating over δ from 0 to 1 the system takes the form

$$\frac{da}{dt} = -\frac{a}{\omega}\frac{d\omega}{dt} + \frac{1}{2}\frac{a}{h}\frac{dh}{dt} + \frac{2}{5}a^2\omega - \frac{4}{5}b^2\omega - \frac{3\nu a}{h^2} + 2b\omega - \frac{3}{2}\omega; \quad (7.1)$$

$$\frac{db}{dt} = -\frac{b}{\omega}\frac{d\omega}{dt} + \frac{1}{2}\frac{b}{h}\frac{dh}{dt} + \frac{6}{5}ab\omega - \frac{3\nu a}{h^2} - 2a\omega - \frac{3}{2\omega}\frac{d\omega}{dt}; \quad (7.2)$$

$$\frac{dh}{dt} = \frac{4}{3}a\omega h; \quad (7.3)$$

$$dm = Q dt. \quad (7.4)$$

We consider the variation of the liquid mass on the plate in time. Since

$$m = \rho\pi r^2 h,$$

$$\frac{dm}{dt} = 2\pi\rho h r \frac{dr}{dt} + \pi r^2 \rho h \frac{dh}{dt} = Q. \quad (8)$$

By solving this equation with respect to dr/dt we obtain

$$\frac{dr}{dt} = \frac{rQ}{2m} - \frac{r}{2h}\frac{dh}{dt}. \quad (9)$$

The initial conditions arise from the fact that the liquid flows out of the batcher with some velocity that is the initial one for spreading over the plate and the dimensions of the drop are determined by the geometry of the batcher, and therefore for system of equations (8) and (9) they are formulated in the following manner:

$$t = 0:\ a(0) = a_0;\ b(0) = b_0;\ h(0) = h_0;\ r(0) = r_0. \quad (10)$$

The value of a_0 can be determined from the condition of equality of the flow rate through the batcher cross section and the total radial flow at the initial instant:

$$a_0 = -\frac{3Q}{4\pi r_0 h_0 \omega_0}. \quad (11)$$

The value of b_0 is calculated from the vanishing of the absolute tangential velocity components at $z = h$:

$$b_0 = 1. \quad (12)$$

The final system of equations that describes the flow of the liquid over the plate can be represented as

$$\frac{da}{dt} = -\frac{a}{\omega}\frac{d\omega}{dt} + \frac{1}{2}\frac{a}{h}\frac{dh}{dt} + \frac{2}{5}a^2\omega - \frac{4}{5}b^2\omega - \frac{3\nu a}{h^2} + 2b\omega - \frac{3}{2}\omega\,; \tag{13.1}$$

$$\frac{db}{dt} = -\frac{b}{\omega}\frac{d\omega}{dt} + \frac{1}{2}\frac{b}{h}\frac{dh}{dt} + \frac{6}{5}ab\omega - \frac{3\nu b}{h^2} - 2a\omega - \frac{3}{2\omega}\frac{d\omega}{dt}\,; \tag{13.2}$$

$$\frac{dh}{dt} = \frac{4}{3}a\omega h\,; \tag{13.3}$$

$$\frac{dr}{dt} = \frac{rQ}{2m} - \frac{r}{2h}\frac{dh}{dt} \tag{13.4}$$

with the following initial conditions:

$$t = 0:\ a_0 = -\frac{3Q}{4\pi r_0 h_0 \omega_0}\,;\quad b = 1\,;\quad h = h_0\,;\quad r = r_0\,. \tag{14}$$

Analyzing the results given in Fig. 2 shows qualitative agreement of the developed model with the process of flow over a rotating substrate.

In the stage of application we will choose the following criteria to compare different regimes of rotation in application:

symmetry of the flow distribution over the plate;

absence of vortex formation on the plate;

minimum time for covering the plate.

As applied to the model considered, these conditions will be reformulated in the following manner.

We will assess the symmetry of the liquid distribution by the simultaneity of the covering of the substrate (the difference between the times in which the edge is reached in opposite directions of the flow):

$$\Delta t = t_{max} - h_{min}\,. \tag{15}$$

The condition on vortex formation on the plate is the vanishing of the vorticity of a liquid particle. Since the layer height is very small and is much smaller than the drop radius, vortex formation is possible only about the axis. To exclude this, it is necessary to satisfy the condition

$$\frac{d\,(Vr)}{dr} - \frac{dU}{d\theta} = 0\,.$$

After substituting V, U we obtain

$$b = 0\,. \tag{16}$$

Thus, we have obtained criteria for assessing the quality of spreading of the liquid over the plate:

$$\Delta t \to \min\,,\quad b \to \min\,,\quad t_{max} \to \min\,. \tag{17}$$

A comparative analysis of liquid spreading performed by these criteria with different regimes of rotation showed that application at a constant rate close to the rate of formation is the most efficient. Symmetric spreading of the dose with the given method of application is confirmed by high-speed filming. With the method found, normally directed forces have a predominant effect during covering; the liquid particles move from the center to the periphery

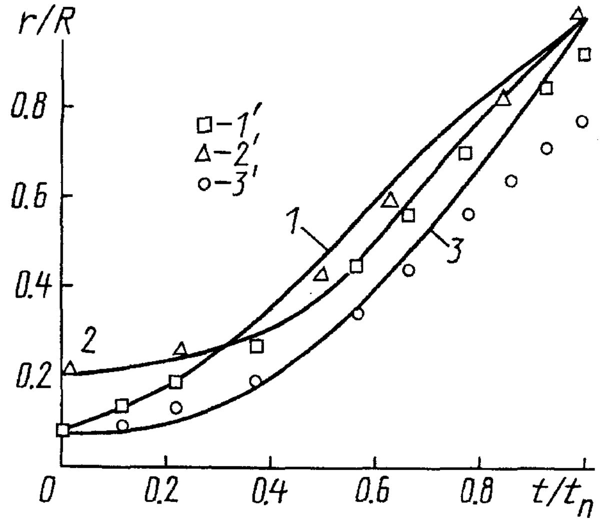

Fig. 2. Variation of the relative radius of the resistor drop in time (t_n is the calculated time of covering of the plate): 1, 1′) values calculated according to the model and experimental values for a liquid of viscosity 26 cSt and for an angular rotational velocity of 5200 rpm; 2, 2′) values calculated according to the model and experimental values for a liquid of viscosity of 42 cSt and for static application and a formation velocity of 4000 rpm; 3, 3′) values calculated according to the model and experimental values for a liquid of viscosity 4.5 cSt and for an angular rotational velocity of 5600 rpm.

of the plate on a radius without vortex formation. No zones of predominant flow are formed. Covering occurs in all directions practically simultaneously, which enables us to decrease substantially the volume of the applied liquid.

Flow in the second stage of the process, i.e., discharge of excesses, was analyzed with the aim of determining the layer thickness at which the flow will cease, the final coating thickness, and the rotation regime for the plate that gives the minimum nonuniformity of the coating produced.

The system of equations that describes flow in the second stage reduces to

$$\frac{da}{dt} = -\frac{a}{\omega}\frac{d\omega}{dt} + \frac{1}{2}\frac{a}{h}\frac{dh}{dt} + \frac{2}{5}a^2\omega - \frac{3\nu a}{h^2} + 2b\omega - \frac{3}{2}\omega\,; \tag{18.1}$$

$$\frac{dh}{dt} = \frac{4}{3}a\omega h\,. \tag{18.2}$$

It describes the flow of a Newtonian liquid. As the layer dries, its properties differ increasingly from the properties of a Newtonian liquid and the validity of the given equations is violated. They are valid only up to a certain moment, namely, the moment when flow ceases, which we determine in the following way. Let the covering time be no more than T for a plate of radius R; then the average velocity of the flow of particles is R/T. The flow can be considered completed at a velocity U that is no more than 0.5% of R/T:

$$U = 0.005R/T\,, \tag{19}$$

but since on the layer surface $U = -a\omega r$,

$$a^* = -\frac{0.005R}{\omega r T}\,. \tag{20}$$

From this equation it follows that the value of the parameter a^* at the moment when flow ceases will be determined not only by the angular rotational velocity but also by the position of the point on the plate.

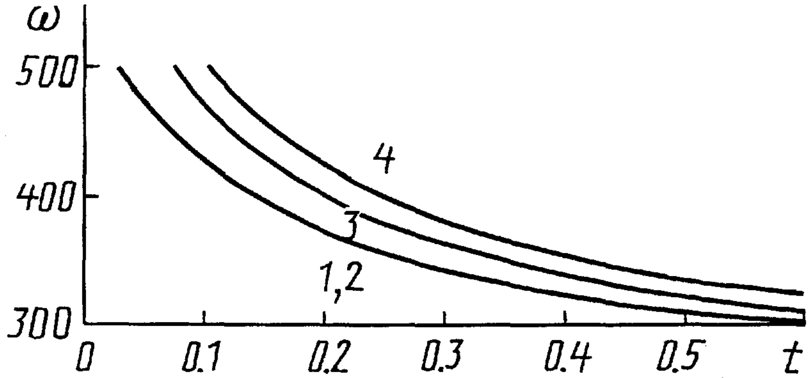

Fig. 3. Calculated regime of rotation of the plate (time variation of the angular velocity): 1) $\nu = 2.2$ cSt; 2) 10; 3) 30; 4) 42. ω, 1/sec; t, sec.

The final layer thickness can be determined from the condition of equality of the centrifugal force and the viscous force under the condition (20):

$$\nu \frac{d^2U}{dz^2} = \omega^2 r . \tag{21}$$

After rearrangements we obtain

$$h = \sqrt{\left(- \frac{a^* \nu}{\omega} \right)} . \tag{22}$$

But the finished coating consists only of the solid component, and the solvent evaporates. To take this factor into account, it is necessary to introduce into Eq. (2.27) a coefficient that takes account of the solid residue content:

$$h = \gamma \sqrt{\left(- \frac{a^* \nu}{\omega} \right)} . \tag{23}$$

Thus, it is established that the final thickness of the coating produced is determined not only by the liquid viscosity, the angular rotational velocity of the plate in the formation stage, and the solid residue content but also by the parameter a^*. And since flow does not cease on the substrate simultaneously, the conditions of formation of the resultant profile vary in time, which leads to the emergence of radial nonuniformity in the layer. However, the analysis performed enables us to determine a regime eliminating this dependence. For this purpose we should satisfy the condition

$$\frac{d}{dt} \left[- \frac{0.005R}{\omega r T} \right] = 0 . \tag{24}$$

After rearrangements,

$$\frac{d\omega}{dt} = \frac{2}{3} a \omega^2 . \tag{25}$$

We have obtained a law of variation of the angular rotational velocity of the plate in time that enables us to produce the most uniform coating and to eliminate radial nonuniformity formed in the second stage of the process, i.e., discharge of excesses.

To calculate the flow of the liquid according to the law derived, it is necessary to use the following system of equations:

$$\frac{da}{dt} = -\frac{a}{\omega}\frac{d\omega}{dt} + \frac{1}{2}\frac{a}{h}\frac{dh}{dt} + \frac{2}{5}a^2\omega - \frac{3\nu a}{h^2} + 2b\omega - \frac{3}{2}\omega ; \quad (26.1)$$

$$\frac{dh}{dt} = \frac{4}{3}a\omega h ; \quad (26.2)$$

$$\frac{d\omega}{dt} = \frac{2}{3}a\omega^2 . \quad (26.3)$$

Figure 3 gives calculated dependences of the variation of the angular velocity. The mathematical model obtained and the analysis performed enable us to draw the following conclusions:

coatings should be formed with a time-varying angular rotational velocity;

the rotational velocity should be maintained constant in the stage of covering of the substrate and be decreased according to the law (26) in discharging the excesses;

the boundaries of the stages and the law of variation of the angular rotational velocity of the substrate are determined by the previous flow of the liquid.

NOTATION

R, plate radius; ω, angular rotational velocity of the plate; Q, flow rate of the incompressible liquid supplied to the plate; U, V, W, radial, azimuthal, and normal velocity components of the volume element of the liquid; ν, kinematic viscosity factor of the liquid; r, θ, z, cylindrical coordinates (the center coincides with the center of rotation); h, m, height and mass of the liquid dose on the plate; a, b, dimensionless functions of time; $\delta = z/h$, relative height of the volume element in a drop of the liquid; t, time; T, time of covering of the plate; γ, solid residue content.

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